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Representative Volume Size: A Comparison of Statistical Continuum Mechanics and Statistical Physics

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Abstract

In this combination background and position paper, we argue that careful work is needed to develop accurate methods for relating the results of fine-scale numerical simulations of material processes to meaningful values of macroscopic properties for use in constitutive models suitable for finite element solid mechanics simulations. To provide a definite context for this discussion, the problem is couched in terms of the lack of general objective criteria for identifying the size of the representative volume (RV) of a material. The objective of this report is to lay out at least the beginnings of an approach for applying results and methods from statistical physics to develop concepts and tools necessary for determining the RV size, as well as alternatives to RV volume-averaging for situations in which the RV is unmanageably large. The background necessary to understand the pertinent issues and statistical physics concepts is presented.

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Acronyms

BC	Boundary Condition
BCI	Boundary Condition Independence
EMT	Effective Medium Theory
FEM	Finite Element Method
RV	Representative Volume
SCM	Statistical Continuum Mechanics

Executive Summary

Brief summaries are presented of effective medium theory for elastic composites and the statistical physics topics of critical phenomena and percolation models. These provide background for a discussion of determining the size of the representative volume (RV) in composites. The aim is to determine *inelastic* effective properties to use in macroscopic material models by averaging over the RV. This method of homogenization of a heterogeneous continuum is just one instance of length scale bridging. By making an analogy between statistical continuum mechanics and statistical physics, we identify a principle on which to base determination of the RV size for inelastic effective properties - namely, that effective property values determined as volume averages over an RV are nearly independent of the boundary conditions (BCs) on the RV. (For the volume averages to be meaningful, the BCs considered are required to be macroscopically uniform.) This principle implies a direct, trial-and-error method for determining the RV size once the field quantity is identified whose average provides an inelastic effective property of interest. We go on to explain the notion of correlation length because its determination presents the possibility for reducing the trial-and-error search for the RV size by a single direct calculation. Numerical investigations to test the ideas developed here are outlined at the end of this report.

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Preface

The overall objective of our investigations of the concepts and tools of equilibrium and nonequilibrium statistical physics is to develop techniques and understanding that will allow us to address problems encountered in modeling the *macroscopic* thermo-mechanical behavior of materials that are heterogeneous on the *mesoscopic* scale, a fine scale continuum that is still much larger than the atomic scale. The application for such material models is in numerical solid mechanics simulations for which the spatial discretization length is larger than the mesoscopic scale.

Bridging the length and time scales between mesoscopic properties and processes and macroscopic simulations is an important step in the development of predictive computational solid mechanics simulations. Among the material modeling problems that our work may help with is developing fracture criteria, which are the rules governing the magnitude and direction of the incremental growth of a macroscopic crack in a solid mechanics simulation. Identifying fracture criteria that are based on mesoscopic (microstructural scale) thermo-mechanical mechanisms is a key problem in developing predictive solid mechanics simulations.

The present inquiry is preliminary work concerned with several generalities related to bridging between scales and with the basis of a common method for linking mesoscopic processes to macroscopic behavior - namely, averaging the mesoscopic details over a representative volume. Accordingly, there is little mention of fracture in this report. Nevertheless, the region near the tip of a macroscopic crack is inherently a nonrepresentative volume. Our consideration of what can be done when the representative volume is too large is preliminary to addressing in the future how to treat the crack tip region at the macroscopic scale. Beyond this we note that percolation theory recently has been extensively investigated as a model for fracture in disordered materials. That a percolation model might reveal aspects of the collective behavior of microcracks makes the current project's pursuit of concepts from statistical physics and critical phenomena an additional contribution to the anticipated follow-on investigation of cracking.

1. Introduction

There is increasing interest within Sandia National Laboratories (SNL) and throughout the DOE weapons laboratories to develop numerical simulation capabilities that can substantially augment laboratory testing and can be used in assessing device reliability. In this context it is important to appreciate that the macroscopic quantities required to predict thermomechanical material response cannot always be calculated accurately at the macroscopic continuum scale. In the absence of a sufficient quantity and variety of experiments, such quantities need to be calculated by directly simulating the micromechanical processes that underlie them. Using micromechanical simulations to determine values of macroscopic quantities requires bridging - *i.e.*, rigorously relating - their disparate length and time scales. Classical approaches for bridging length scales posit the existence of a "**representative volume**" (RV) - a volume over which simple volume averaging of the properties or responses of the constituent phases provides a useful description of the overall thermo-mechanical response of the heterogeneous material. This motivates the question: "*How can the size of the RV be determined for a material whose macroscopic response is calculated by simulating micromechanical processes?"* A natural subsequent question is then: "*How can the results of microstructural scale simulations be related to macroscopic quantities when volume averages over an RV are not feasible?"* The purpose of this report is to review the basic results from **effective medium theory** (EMT), the theory of determining characteristic overall properties of composites (heterogeneous continua) and discuss fundamental similarities of EMT to the modern view of statistical mechanics. These both contribute to the most basic purpose, that of identifying an approach for developing a direct means of determining the RV size. For clarity the term "**statistical physics**" will be used instead of "statistical mechanics" to refer to the averaging of microphysics phenomena. This will distinguish it from "**statistical continuum mechanics**" (SCM), which refers to the averaging of micromechanics phenomena. Though much of the discussion is general, our intended application is to numerical simulation of the thermomechanical response of solids.

(A note on terminology: Micromechanical processes are those occurring on the microstructural scale, which is finer than the homogenized continuum or finite element mesh resolution but is large enough to be treated as a heterogeneous continuum. Here "microstructure" refers to any heterogeneities in the continuum, not only to the crystal grain structure. It is usual that microstructural scales in continua are much larger than atomic scales. For this reason these scales are often referred to as "*mesoscopic*" rather than "microscopic.")

The application of interest to SNL that concerns us here is using numerical simulations of microstructural-level processes to generate synthetic constitutive data. The simulated data provide the basis for constructing or extending a macroscopic material model that can be used in standard **finite element method** (FEM) simulations of solid mechanics. Restated, we seek to use the numerical simulation results to determine the equilibrium, macroscopic thermo-mechanical properties of a *homogeneous effective medium* whose response is the same as the RV-averaged response of the simulated material. (The additional difficulties of determining nonequilibrium quantities are commented on later.) As explained below, it is only for regions as large or larger than the RV that an EMT can provide an accurate description of macroscopic thermo-mechanical response. Adopting a numerical approach to homogenization largely obviates assumptions about the distribution of

heterogeneities and the nature of their interactions because it is able to treat a wide range of realistic distributions and interactions. This capability is the main advantage of determining bulk material properties from mesoscopic numerical simulations. However, this versatility also makes identifying the size of the RV a *necessity*: The simulated system must be as large as the RV for the simulation results to provide overall responses that are representative of macroscopic constitutive behavior. Because the simulated materials will be heterogeneous on the mesoscopic scale, they will be referred to, in general, as *composites*.

Given that homogenized descriptions of the response of heterogeneous materials are commonly used in continuum mechanics, it is worth explaining why RV size in nonperiodic materials is rarely determined, if ever. A typical SCM treatment defines densities as volume averages over a hypothetical but unspecified RV. These densities are then used as though they were truly thermodynamic quantities. This type of micromechanics development is carried out without regard to the size of the RV on the basis of assuming a uniform distribution of heterogeneities and that their mutual interactions are either negligible (dilute concentration case) or self-consistent (mean-field case). The resulting theoretical prediction is internally consistent, but its validity is limited to regions equal in size to the RV or larger. Hence determining the RV size should actually be part of the *application* of this type of SCM result. Situations intermediate to these limiting cases are of greatest concern for specific determination of the RV. That the RV size is not determined for these applications points out the potential danger that an RV-based method can be applied inappropriately when the RV is larger than the macroscopic volume of interest, if it exists at all. Without explicitly identifying the RV size, it may not be evident that a classical scale-bridging method is without a sound basis and that predictions based on the resulting material model are incorrect. This unsatisfactory situation exists because there is no objective definition or set of criteria for establishing that a chosen averaging volume in a nonperiodic material is actually an RV.[1]

Relating the mesoscopic details of geometry, properties, and deformation processes to average mechanical properties of a macroscopically heterogeneous material is a central pursuit of statistical continuum mechanics (SCM). This averaging problem appears highly analogous to the averaging problem of statistical physics. Here we use statistical physics as a guide to clarifying the meaning of macroscopic effective properties of a heterogeneous continuum. Whether statistical physics also provides a guide to determining values of macroscopic effective properties of composites requires further comparison and consideration, which is begun in this report. Identifying similarities between statistical physics and SCM is useful for providing a unified conceptual framework in which to view the two averaging problems. It is hoped that this will be of additional use in leading to practical solution procedures in one or both of the disciplines, based on approaches used in the other.

The ultimate goal of statistical physics is understanding how continuum physics is a manifestation of the behavior of a dynamic system of a very large number of microscopic, interacting particles occupying a volume very much larger than the particle dimensions. In particular, equilibrium statistical physics determines how to compute the densities of macroscopic mechanical quantities - i.e. any extensive quantity per unit volume - that appear in equilibrium thermodynamics theory as averages over the motions in a system of many atomic-scale particles. This provides the justification of equilibrium thermodynamic theory

for homogeneous systems as well as for macroscopically heterogeneous systems that are not too far from equilibrium. A heterogeneous body is regarded as comprising many small, macroscopic volumes that are homogeneous and in equilibrium with the local conditions. In each small volume equilibrium thermodynamics is taken to govern the densities of thermodynamic variables. The overall behavior of the macroscopic body is given by macroscopic phenomenological dynamical relations that describe the changes of the thermodynamic densities. Examples are reaction-diffusion equations and continuum momentum flux equations. The dynamical behavior of such a heterogeneous body is assumed not to violate the underlying assumptions of local thermodynamic equilibrium.

The application of statistical physics to describe *nonequilibrium* macroscopic processes - i.e., to derive the continuum equations of motion from the dynamics of a system of many particles - is an area of current research. Being able to describe nonequilibrium processes is a goal of our investigations, but it will take much effort to accomplish. While some discussion is given below of the additional complications that occur when treating nonequilibrium phenomena, the scope of this report is largely limited to equilibrium properties.

In the next section the RV is defined as the minimum volume of material for which the overall properties are independent of boundary conditions. **Boundary condition independence** (BCI), the insensitivity of average property values to the choice of allowed BCs, is argued to be appropriate and sufficient for defining the RV for time-independent properties because it is the essential aspect of what is meant by material property. BCI provides a conceptual link to bulk property determination in statistical physics. This connection is discussed in the third section. In Section 4 key results of EMT are summarized and the implied formal procedure for identifying the RV size is given. This leads into the discussion of establishing a practical procedure for identifying the RV size in Section 5. There it is noted that BCI should be strongly influenced by the geometrical configuration of the heterogeneities in the volume. Thus the task is to identify which aspect of the internal geometry is key and how to determine when it reaches a state that provides BCI. Because of its physical significance, the correlation length is a low-order measure of a statistical distribution that is a potentially useful quantity to use to express the criteria that the internal geometry must meet for volume averages to be BC independent. To illustrate its physical meaning, basic aspects of critical phenomena and the theory describing it are presented in Section 6. Brief descriptions of the scaling hypothesis and percolation theory are also presented in this section along with a discussion of the possible relevance of critical phenomena to micromechanical modeling. The initial study that will be pursued to begin investigating the issues and possibilities raised in this report is outlined in Section 7.

2. Boundary Condition Independence of RVs

We begin with Hill's characterization of a representative volume:[2]

> "This phrase will be used when referring to a sample that (a) is structurally entirely typical of the whole mixture on average, and (b) contains a sufficient number of inclusions for the apparent overall moduli to be effectively independent of the surface values of traction and displacement, so long as

> these values are 'macroscopically uniform.' That is, they fluctuate about a mean with a wavelength small compared with the dimensions of the sample, and the effects of such fluctuations become insignificant within a few wave-lengths of the surface. The contributions of this surface layer to any average can be made negligible by taking the sample large enough."

It is understood that the volume-averaged ("overall") properties of the RV are the desired effective properties of the hypothetical equivalent homogeneous material. Hill's characterization is notable both for being one of the first and for being possibly the only one that includes near independence of the average properties from the boundary conditions [part (b)] as fundamental to the RV. For the two reasons that follow, we take the sole defining feature of the RV to be the independence (to some prescribed precision) of the volume average properties from the boundary conditions. First, BCI is the essence of what is meant by "material property." In application the properties of a material are taken to have values that are fixed or possibly depend on local mechanical field variables, and these values can be used to calculate the response of a sample of that material to any possible boundary conditions. Aspects of a specimen that do depend on boundary conditions are structural properties, not material properties. Second, the structural similarity referred to in part (a) of Hill's characterization is both unneeded and overly restrictive for defining the RV, as explained next.

Several other workers who have made substantial contributions to the averaging problem of SCM hold part (a) of Hill's characterization, alone, to be the defining feature of an RV; However, they elaborate on it further[1,3-7]or make due with an imprecise characterization.[8] The structural similarity of the RV to an arbitrarily large volume of the composite is referred to as **statistical uniformity**[1] or **statistical homogeneity**.[8] It is an undesirable constraint because it is a restriction on the composite material as a whole as well as on the RV. If a composite is statistically homogeneous, then this characterization implies that the RV can be identified as the minimum volume for which the statistical description of the geometrical heterogeneity is, for practical purposes, indistinguishable from that of a very much larger sample of the composite. This implication is clear, and it suggests an approach for identifying the RV is to evaluate the variation of some measure of the heterogeneity with sample size. However, as discussed in the introduction, the RV size is never actually determined in theoretical treatments. One reason statistical homogeneity is often required in theoretical developments of effective properties is that when it holds, volume averages over the RV, or any larger volume, equal ensemble averages. The equality of these averages, called **ergodicity**, allows the tools of ensemble theory from statistical physics to be applied to the problem of determining effective material properties of composites.[1,5,7]

(This notion of ergodicity is an analog of the notion originally developed in statistical physics, where the ergodic hypothesis asserts the equivalence of time averages and ensemble averages.[9] Ergodicity in statistical physics was long believed to be necessary for a microphysics system to achieve thermodynamic equilibrium, which is a special kind of statistical uniformity. This is why the concept of ergodicity is appealing in the problem of effective properties in SCM. In this light it is interesting to note that in modern statistical physics, it is understood that ergodicity is not prerequisite for the establishment of thermo-

dynamic equilibrium. Conservative Hamiltonian systems that are fully integrable can be ergodic.[10,11] It is now understood that thermodynamic equilibrium and macroscopic irreversibility arise from the phase space mixing behavior of nonintegrable conservative Hamiltonian systems.[10,11] Mixing, in turn, arises from deterministic mechanics in systems that exhibit a sensitive dependence on initial conditions and, consequently, tend to become chaotic. So it is that the analogy to statistical physics used to treat the averaging problem in SCM has become outdated. It would be interesting to consider whether a new, more useful analogy might be developed based on current statistical physics understanding.

Statistical homogeneity is unneeded because it is not essential to the meaning of material property, and it can be dispensed with because it is unrelated to BCI. The value of a material property can vary over macroscopic distances and still be well-defined locally (on the macroscopic scale), according to the criterion of BCI. Statistical homogeneity constrains the value of a material property from having any spatial variation. This constraint is clearly not essential to the meaning of effective material property. Indeed it is unwanted restriction on what is meant by effective material property. In addition we note that ensembles are merely a tool in statistical physics that does not add to our understanding of how fine-scale properties and processes are manifest in overall response.[10] Ensemble averages have no strict meaning in any one sample;[1,10] yet we need to treat individual samples if we are to understand the relation of the fine scale to macroscopic properties in both statistical physics[10] and SCM.

Hill's restriction to macroscopically uniform boundary conditions deserves comment regarding both the *determination* and the *use* of effective material properties. First, macroscopically uniform loading is typically used in determining material properties, even in homogeneous materials, because their use simplifies relating the measured response of a specimen to a material property. Also it seems reasonable to expect it to be possible to obtain BC-independent overall properties only for macroscopically uniform loadings. Second, the implied restriction to using effective properties only in describing the response to macroscopically uniform loading is easily understood by analogy with the "material point" of the continuum in an actual homogeneous material. The RV should be understood to be the smallest volume of the composite material that can be treated as a homogeneous continuum. In this sense, the RV is analogous to the continuum material point, which is the smallest volume of the material that can be regarded as a continuum. As such all features of the continuum material point, the material property values and the field variables, must be spatially uniform (*cf.* Ref. 12, p. 1; Ref. 8, p. 38). The continuum description is accurate only when the loading produces no significant gradients over the dimensions of the continuum material point.

For example, the homogeneous material cannot be treated as a continuum to describe its response to very high frequency vibration. Accurately describing this loading involves the vibrational modes of the discrete lattice. Similarly the response of a body composed of a composite material can be accurately described as though it were composed of the homogenized material only if the mechanical fields are uniform over volumes the size of the RV. This is achieved by loading that is macroscopically uniform over distances comparable to the linear dimension of the RV. That is, effective property values can be used to describe

the response of a body composed of the heterogeneous composite only when the loading is sensibly constant over distances comparable to the linear dimension of the RV. Though spatial variation of effective property values needs to be accommodated, the effective properties should be nearly constant within regions comparable to the RV size for the response to be accurately described by that of a nonuniform homogenized material. So it is useful and appropriate to regard the RV as a very large continuum material point for the fictitious, homogenized material. The requirement for macroscopically uniform BCs restricts the allowed BCs to the specification of a uniform traction or displacement vector on each pair of coordinate faces of the body.

Next we ask the question, "Should we expect that an RV exists?" In other words, should we expect that for a sufficiently large volume, the overall properties of the material become nearly independent of boundary conditions? We know that RVs exist for some materials. For example, fine grained, polycrystalline, structural metals are well described as homogeneous, isotropic materials for many practical applications; but, in general, the existence of an RV is an empirical question. It can only be answered by observing the behavior of a composite material under the relevant conditions. Furthermore, we should anticipate that whether an RV exists and its particular size are dependent both on the property of interest and the accuracy required by the application at hand. Nonetheless, it may be possible to enumerate classes of micromechanical behavior that are compatible with existence of an RV.

For a material that has an RV, we ask: "What is it that makes the overall properties of the RV BC independent, whereas those of any smaller volume are not?" The contrast in property values of the constituents of a composite has been demonstrated by numerical simulation to effect the RV size of an elastic composite.[13] We expect this dependence to persist for inelastic composites response as well, but for a given composite, we presume that the effect of the contrast in the constituent's property values only determines a scale factor. The property value contrast is thus assumed to be a parameter and, consequently, BCI in a given composite is taken to only involve aspects of the statistical description of the internal geometry the shapes, configuration, and spatial distribution of the heterogeneities. This assumption provides some guidance for developing a direct method of determining the RV size, rather than a trial-and-error search for a volume that provides nearly BC-independent average values. This is pursued further in Section 5.

3. Bulk Properties in Statistical Physics

3.1 Macroscopic Equivalence

In applying statistical physics to understand most continuum phenomena, the task amounts to evaluating the bulk properties of a material.[14] Based on the observation of locality (also known as "local action") in macroscopic continua - *i.e.*, that the values of densities at a given spatial location are determined solely by the environment at that location - Balescu argues for adopting locality as a principle of statistical physics theory.[14] This principle is equivalent to requiring that properties of the macroscopic system be *independent of*

boundary conditions. Formally, this is expressed by the **principle of macroscopic equivalence**.[14] Consider the value, λ , of some material property **density** - *i.e.*, a material quantity per unit volume - for a sequence of system volumes, $\{v_k\}$, that increases without bound. The members of the sequence are successively enlarged systems that leave unchanged the *local conditions* at equivalent locations within the different systems. Requiring macroscopic equivalence restricts the system size dependence of the intensive quantity, λ_k, to have the form

$$\lambda_k(v_k) = \Lambda + \hat{\lambda}_k(v_k) . \quad \textbf{(1)}$$

Λ is the bulk value common to any macroscopic system in the class, provided it is not too small. The remaining contribution to λ has a volume dependence arising from the influence of the system boundaries. Because this source of size dependence decreases as the surface-to-volume ratio decreases, its volume dependence has the limiting behavior

$$\hat{\lambda}_k(v_k) \to 0 \text{ as } v_k \to \infty . \quad \textbf{(2)}$$

The systems having a common bulk value of a property are said to be "macroscopically equivalent." The goal is to determine the bulk value, Λ. Once it is known, it is the appropriate value of λ to use to describe this material property for any of the macroscopically equivalent systems. Let $k = \kappa$ denote the value of the index for which λ_k is sufficiently close to Λ. This means that we do not have to specifically treat the correction, $\hat{\lambda}_\kappa(v_\kappa)$, arising from BCs because the system volume is large enough that the surface effects make little contribution to the value of the intensive quantity. This is to say that according to Eqs. (1) and (2), which are consequences of the principle of macroscopic equivalence, the removal of size effects corresponds to BCI.

The bulk value is often most amenable to determination in the physically unobtainable **thermodynamic limit** because it automatically removes boundary effects so that $\lambda_\kappa \approx \Lambda$. This is the limit in which the size and number of particles in a system become arbitrarily large while maintaining a constant particle concentration:[9,14]

$$v \to \infty \text{, and } N \to \infty \text{ with } \frac{N}{v} = \text{ constant.} \quad \textbf{(3)}$$

From this description it should be understood that to state that some condition or relation holds "in the thermodynamic limit" is a shorthand way of saying that the condition or relation is satisfied by the BC-independent bulk value of the property or properties under consideration. Unambiguous values or expressions for densities in terms of averages over microscopic quantities are obtained by rigorous mathematical evaluation of the thermodynamic limit. In statistical physics we know that nearly BC-independent averages exist as these are simply the macroscopic mechanical properties of the homogeneous materials that are described by thermodynamics. Not all quantities fit this description. Thermodynamics

is also concerned with thermal quantities which are related to the state of a system rather than to averages of microphysics variables.[14] Also macroscopic properties do not exist for all systems.[14] Balescu gives as examples thin films, for which one dimension is not macroscopic, and ultradilute gases, in which nearly all particle collisions are with the container walls so the entire macroscopic behavior is due to boundary conditions alone.

There is one other size dependence to λ in addition to that arising from surface effects. This is that the system be sufficiently large for Λ to have the same value as any larger macroscopically equivalent system. To understand the cause of this size dependence, consider a practical determination of the value of Λ for a finite-sized system. Namely, take Λ to be the average of the quantity, λ, over a region in the interior of the system that is sufficiently distant from the surfaces to be insensitive to the BC on the system. The value of Λ determined in this way will depend on the size of the region over which the average is taken until the minimum linear dimension of this region exceeds the correlation length of the bulk system. As explained below in Section 6, the correlation length, ξ, is the characteristic length of the bulk system. It is a measure of the range over which the response in one portion of the system is strongly related to that in another portion of the system. A size-independent bulk value is not obtained until the averaging region is sufficiently larger than ξ that it samples largely independent portions of the bulk system. In Section 6 it is also shown that a correlation length characterizes the range of influence of the surface conditions. This correlation length possibly is a different one than for the bulk system.

The expression for stress in a system of discrete particles provides an example of several of the points made in this section. This is presented in the appendix.

For nonequilibrium systems the concepts presented above are applied in the following way. The locality principle is taken to be that the rates of change of the densities depend only on the local conditions. Equations (1) and (2) are assumed to be true for times less than some characteristic time, which is taken to increase without bound as the volume increases.[14] Finally, the analog of the thermodynamic limit for nonequilibrium systems is called the **hydrodynamic limit**;[15] however, it applies equally to solids and fluids. The hydrodynamic limit takes the time scale into consideration and is much more complicated. It is the subject of highly technical current research.

3.2 Comparison of Statistical Physics and SCM

It is interesting that BCI is the essential requirement for determining bulk properties in both statistical physics and SCM. However, this is in part by design. The statistical physics explanation of what is meant by “macroscopic bulk property” provides the framework for characterizing the relation of the properties of an effectively homogeneous material to those of an underlying heterogeneous medium. Thus, the arguments in Section 2 supporting taking BCI as the defining feature of the RV were guided by the statistical physics description of the bulk value of a material property. The similarities and differences need to be considered further if we are to learn something from the comparison of the averaging problems in SCM and statistical physics.

The formal description of the bulk value of a material property for a macroscopically large microphysics system given by Eqs. (1) and (2) at first appears incompatible with the notion of effective properties of a heterogeneous continuum developed in Section 2. While Balescu does require a minimum system size,[14] this does not seem to be the RV in the sense of Section 2 because the BC-independent bulk value is formally achieved only in the infinite volume limit and not for the minimum allowed system size. (Indeed, Balescu's intention in setting a minimum size requirement for Λ is not entirely clear.) Making an explicit analogy from the statistical physics description given by Eqs. (1) and (2) to SCM might lead one to conclude that BC-independent averages should not be expected for any volume of composite but that the bulk value should be determined in the fictitious infinite volume limit and then used as the value for the material property for finite composite systems.

As a practical matter the discrepancy between the two disciplines is not so severe. When the sequence of systems in Eq. (1) converges quickly, as is often the case,[14] surface effects can already be negligible for Balescu's minimum size for a macroscopically equivalent system. In this case the minimum-sized, macroscopically equivalent system can be consistently interpreted to be the continuum material point. Consequently, the system size at which the material property is equal to the bulk value, within the desired precision, is completely analogous to the RV in SCM as characterized in Section 2.

This discussion identifies a sense in which the statistical physics and SCM characterizations of material properties are compatible. As a consequence, it suggests that the bulk value or effective property value can be obtained by considering the fictitious infinite volume case and then used as the value of a material property for systems of any size greater than some minimum - namely, the RV. However, it is unlikely to be practical to numerically determine effective properties of a heterogeneous continuum from the infinite-sized system. Moreover, such an approach for a heterogeneous continuum still requires identifying the RV size, which is to be taken as the minimum system size that Balescu stipulates.

Another difference from between the two disciplines is that the statistical physics characterization of macroscopic material properties is more general than the SCM characterization of effective material properties in that it applies equally to any macroscopic field variable. The bulk value is to be taken as the macroscopically local value of a macroscopically nonuniform field. The restriction that the fields have negligible variation over some minimum microscopic volume is not stated[14] but is, perhaps, implicit in the concept of macroscopic fields. This is worth further consideration in the context of specific applications.

Differences between the systems treated in the two disciplines may be critical to establishing an analogy between statistical physics and SCM. In statistical physics systems the linear size of the molecular constituents relative to the macroscopic system is four orders of magnitude for a nanogram of material. Correspondingly macroscopic response times of interest are much longer than subnanosecond atomic equilibration times. In contrast SCM takes microstructural elements as the fine-scale constituent. A small macroscopic volume will not necessarily contain many of the fine-scale constituents nor will macroscopic time intervals of interest necessarily be much longer than characteristic times of some micromechanical processes. Henc, there is a much smaller disparity of length and time scales between the fine scale and macroscopic scale of a heterogeneous continuum than there is for

statistical physics systems. This is expected to have consequences for the accuracy of statistical physics techniques applied to SCM systems. It remains to be determined how severely the accuracy of an effectively homogeneous description of a heterogeneous continuum is reduced as compared to that for a statistical physics system.

A last difference is that the constituents in a statistical physics system are governed by dynamical equations for discrete constituents, and the averages of interest are over a dynamic steady state (for equilibrium). In SCM static force balance governs the fine-scale constituents and the averages are over static geometric configurations. The statistical correlations of interest also differ. For statistical physics the mean free path of the particles may be key, whereas in SCM the important measure may be certain moments of the heterogeneity distribution.

4. Effective Medium Theory and RVs

4.1 The Hill Condition

The goal of effective medium theories (EMT) is to represent the behavior of a heterogeneous material by a homogeneous material whose properties equal the RV-averaged properties of the heterogeneous material. Doing so has the practical advantages of reducing the detail of the material description to only those aspects that are of interest on a large scale and making available for the heterogeneous material all of the numerical and mathematical analysis techniques developed for treating homogeneous bodies.[1,5] Effective properties must be determined in a statistical sense because we do not have complete knowledge of the heterogeneous material, or we want to describe nominally equivalent samples that differ in fine-scale details. Energy principles can be applied to determine theoretical bounds on the properties using whatever information is known about the material's constitution.[5] Alternately an experimental determination of an effective property is made by testing a collection of samples to determine a statistical estimate.[7,16] The resulting bounds or values are then used to calculate the average mechanical response of the heterogeneous material.

Constraints exist on the circumstances for which such an effective medium description is appropriate. The constraints insure that the effective medium behaves like an ordinary continuous medium by requiring that averaged balance equations (mass, momentum, energy, entropy) have the same form as the local balance equations for a uniform continuum.[17] For example, consider the linear elastic response of a polycrystal. We are interested in the effective elastic constants (EEC). The constraints simplify to requiring that the EEC defined energetically and mechanically are compatible. Defined mechanically, the EEC are denoted C^M_{ijkl} and are the proportionality constants between the volume-averaged stress and volume averaged strain tensors,

$$\langle \sigma_{ij} \rangle = C^M_{ijkl} \langle \varepsilon_{kl} \rangle . \quad \textbf{(4)}$$

(Summation is implied on repeated indices.) Defined energetically, they are denoted C^E_{ijkl} and are the proportionality constants between the volume-averaged strain energy and the volume-averaged strains

$$\langle U \rangle = \frac{1}{2} C^E_{ijkl} \langle \varepsilon_{ij} \rangle \langle \varepsilon_{kl} \rangle = \frac{1}{2} \langle \sigma_{kl} \rangle \langle \varepsilon_{kl} \rangle . \quad \textbf{(5)}$$

But $\langle U \rangle$ is just the volume average of the local strain energy

$$\langle U \rangle \equiv \frac{1}{2} \langle \sigma_{ij} \varepsilon_{ij} \rangle = \frac{1}{2} \langle \sigma_{ij} \rangle \langle \varepsilon_{ij} \rangle + \frac{1}{2} \langle \sigma'_{ij} \varepsilon'_{ij} \rangle \quad , \quad \textbf{(6)}$$

where the primes denote the fluctuation of the variable about its volume average value. Substituting for $\langle \sigma_{ij} \rangle$ from Eq. (4) and equating to Eq. (5) yields

$$(C^E_{ijkl} - C^M_{ijkl}) \langle \varepsilon_{ij} \rangle \langle \varepsilon_{kl} \rangle = \langle \sigma'_{kl} \varepsilon'_{kl} \rangle . \quad \textbf{(7)}$$

This result shows that the mechanical and energetic definitions of the EEC are compatible *only if* the heterogeneous material is loaded so that the volume average of the product of the stress and strain fluctuations (the covariance) vanishes

$$\langle \sigma'_{ij} \varepsilon'_{ij} \rangle = 0 . \quad \textbf{(8)}$$

This constraint, first determined by Hill,[2] expresses the requirement that the external forces deforming the polycrystal are not correlated with its microstructure.[5] For an arbitrary volume of a crack-free heterogeneous material, the Hill condition, Eq. (8), is satisfied by uniform applied static tractions or linearly varying displacements.[2] This class of equilibrium applied loads will be referred to as uniform boundary conditions (UBC). Equation (8) is also satisfied by selected macroscopically uniform mixed boundary conditions.[18-19] The Hill condition, Eq. (8), has been generalized to arbitrary thermomechanical problems,[17] which makes it useful also in defining nonlinear effective media. The UBC have been similarly generalized.[16] No equivalent results are available for developing effective medium descriptions for dynamic, nonequilibrium systems. One difficulty in developing a dynamic theory is that the inertial forces are correlated with the microstructure so that, in general, Eq. (8) is not satisfied.[5] In practice Eq. (8) cannot be satisfied for many composites of interest. Foremost among these are materials containing microcracks. As discussed below, the common practice is to set bounds on energetically defined effective properties or evaluate mechanically defined effective property values from an explicit micromechanical model.

The Hill condition imposes a constraint on the kinds of macroscopically uniform loading that are appropriate in determining effective property values or using them to describe the macroscopic response of a composite. It does not provide BCI. For BCs that satisfy the Hill condition, the EECs determined from Eq. (4), by applying a set of displacement UBCs,

will be compatible with those determined from Eq. (5). However, the resulting EEC tensor will not necessarily be the inverse of the effective compliance tensor determined from, say, the mechanical definition

$$\langle \varepsilon_{ij} \rangle = S^{M}_{ijkl} \langle \sigma_{kl} \rangle , \tag{9}$$

by applying a set of traction UBCs. The different boundary conditions lead to different values of the effective properties. It is implicit in Hill's characterization of the RV that this difference owing to BCs is reduced below some prescribed small value when the averaging volume is an RV. There is some ambiguity in the literature on this point,[1,7] but this conclusion is supported by other theoretical developments[5,8] and illustrated by numerical simulations of elastic composites.[13,19] This property of the RV to provide averages that are BC independent is sometimes called "statistically representative,"[8] which should not be confused with the property that the effective properties are independent of location within a large body, which is called "statistical homogeneity."

This discussion suggests the following procedure for identifying the size of the RV of a micromechanically simulated material exhibiting regular (noncritical) behavior. The size of the RV can be chosen to be the volume whose volume-averaged response yields effective properties that are independent of the UBC within some prescribed tolerance. Establishing a practical, direct method for identifying the RV, instead of this trial-and-error procedure, is discussed in the next section.

Boundary condition independence does not depend on satisfying the Hill condition, Eq. (8). Even though the effective properties defined mechanically and energetically may not be compatible, when BCI is achieved, we have, to within some prescribed tolerance, $S^{M}_{ijkl} \approx (C^{M}_{ijkl})^{-1}$ and $S^{E}_{ijkl} \approx (C^{E}_{ijkl})^{-1}$. S^{E}_{ijkl} is the effective compliance tensor determined from the energy expression corresponding to Eq. (5).

$$\langle U \rangle = \frac{1}{2} S^{E}_{ijkl} \langle \sigma_{ij} \rangle \langle \sigma_{kl} \rangle . \tag{10}$$

In these circumstances the average elastic strain energy also is BC-independent, which corresponds to the merging of the bounds on the effective properties that are provided by energy principles.[8] Finally, when averaged over the RV or any larger, fixed volume at a fixed location, Eq. (7) shows that the volume averaged stress-strain covariance becomes

$$\langle \sigma'_{kl} \varepsilon'_{kl} \rangle = \langle \varepsilon_{ij} \rangle K_{ijkl} \langle \varepsilon_{kl} \rangle , \tag{11}$$

where K_{ijkl} is a constant tensor.

As noted above, the Hill condition cannot be satisfied for many composites of interest. Materials containing cracks stand out prominently among such composites. Whether the effective properties of such composites are defined mechanically or energetically, the RV can still be identified as the volume for which they become sufficiently independent of BCs.

The incompatibility of the mechanically and energetically defined effective properties formally constrains their use to only constitutive calculations or energy calculations, respectively. The advantage of choosing to work exclusively with energetically defined effective properties is that bounds can be put on the values by applying energy extremum principles.[3,8] Use of energetically defined effective properties in macroscopic material models is probably an acceptable approximation when only bounds on effective property values are known. The other common approach, referred to in the Introduction, is to make enough assumptions to sufficiently specify the distribution of heterogeneities within the RV and their mutual interactions. Then an explicit micromechanical description of the behavior of the constituents is developed. The overall behavior is determined by first solving for the response of one constituent to chosen applied BCs and mutual interactions. This response is then averaged over the RV. Because the distribution of the interacting constituents was specified, the RV average is rendered by averaging the single constituent response over this distribution. In this way mechanically defined effective properties are obtained. The resulting analytical expressions, not bounds, for the effective properties are formally restricted to being used in constitutive descriptions and then only for those volumes larger than the RV within which the assumed distribution and interactions obtain.

It should be evident from the discussion of the RV in Section 2 and the limitations of effective medium theories in the present section, that it is highly unlikely that a viable RV can be identified near the tip of a macroscopic crack. This is primarily because the loading of any region near the crack tip has large gradients and so is not macroscopically uniform. Although if the response of a system of microcracks can be well modeled as critical behavior (Section 6), characteristic - *i.e.*, critical - patterning and coalescence of a population of microcracks might be determined.

We have presented two basic theoretical results from EMT. First, the requirement for vanishing stress-strain covariance, Eq. (8), in order for mechanically and energetically defined effective properties to be compatible. Second and independent of the compatibility issue, the empirically based expectation that as the averaging volume is increased, effective properties tend to become BC independent and, correspondingly, the energetic bounds on their values merge. This expectation should, presumably, be expressible as reasonable assumptions about the volume dependence of the heterogeneity distribution of a composite. (This matter is pursued further in the next section.) Whether BCI actually occurs is an empirical question that is only settled by measuring the response of the composite of interest. Both results from EMT have been demonstrated for composites with elastic constituents subjected to small deformations. These basic concepts and results from EMT are potentially highly relevant to the macroscopic modeling application of interest to SNL that was described in the Introduction. However, before their usefulness can be realized, they need to be extended in the following ways: The range of deformations should extend to finite strain, inelastic constitutive behavior of the constituents of the composite should be allowed, effective inelastic properties need to be determined, numerical methods should be developed for treating the discontinuous strain fields of cracked material with arbitrary crack distribution and interactions. These extensions appear to be achievable. As already noted, generalizations of the Hill conditions for arbitrary thermomechanical problems have been reported.[17] Several results relating average stress or strain to applied loads are applicable for fi-

nite strain and inelastic constituent behavior (see comment, ref. 8, p. 35). Finally, the notion that BCI is the defining quality of effective properties and that apparent properties should tend to BCI values as the averaging volume increases seem to pertain equally, regardless of the particular constitutive behavior of the composite constituents.

4.2 Comparison with Bounds on Effective Property Values

For completeness the differences need to be discussed between the intended application of generating synthetic constitutive data and the more traditional pursuit of determining bounds on effective property values. A full discussion will be given elsewhere, or in a revision of this report, after we acquire a sufficient understanding of variational bounding methods and the interpretation of resulting bounds. Here we present several questions whose answers depend, in part, on an understanding of variational bounding methods.

The goal in attempting to generate synthetic constitutive data from mesoscopic scale simulations of the thermo-mechanical response of sufficiently large systems is to develop a macroscopic description of the behavior of the simulated material in which the material is treated as homogeneous and uniform over some small, but macroscopic, volume. The resulting material model would then be used in numerical solid mechanics simulations to analyze the structural response of macroscopic objects of interest. There are two distinct probabilistic aspects of this procedure to generate synthetic constitutive data. They both arise from the sample-to-sample variation of the exact configuration of heterogeneity in the material. In this sense the probabilistic aspects are two sides of the same coin.

The first probabilistic aspect of the procedure is encountered in identifying the size of the RV and values of the effective properties associated with it. It is necessary to assume that the statistical description of the mesoscopic-scale material heterogeneity can be determined to a practical extent. This statistical description will be expressed in the form of a distribution function, and it will be determined from microscopic observations on a set of samples of the material: an ensemble. The statistical distribution for the ensemble of samples can be taken to describe the heterogeneity in a “typical” sample of the material. Because a “typical” sample may never actually occur, this interpretation of the ensemble distribution function should be likened to a median value rather than a mean value. The RV size and effective property values will be determined for the “typical” example of the material. This is necessary both to make it possible to define the RV and to obtain values that are more representative of the average response of a population of samples.

The extent to which the effective property values are representative depends on two factors. One is how well the material response to a particular loading can be treated as the response of a homogeneous material. The other is how closely the heterogeneity distribution in any individual sample approximates the distribution function for the ensemble. This is the second probabilistic aspect of the procedure. It has the consequence that the macroscopic response determined using effective properties will not match the response of any given sample. This adds the practical necessity of estimating the variance of the effective properties to assess the range of responses that can occur in a population of objects made of the heterogeneous material. From this it is seen that the macroscopic solid mechanics

simulation that was supposed to be enabled by determining effective material properties now needs to be a stochastic analysis to account for the variability of individuals from the "typical" behavior.

Accounting for the uncertainty in the input to an analysis requires an additional measure of error besides the variances of the effective property values. The empirical ensemble distribution function will not exactly represent the total population of material samples. Thus a numerical analysis of an ensemble will provide an ensemble-averaged value and variance for an effective property that can differ from those obtained by laboratory testing a collection of samples, each larger than the RV. The laboratory specimens sample the true distribution function, whereas the numerically generated systems sample the ensemble distribution function, which only approximates the true one. Ignoring measurement error, what can we say about the magnitude of the disagreement between the numerical result and the laboratory result? There is an additional consequence. Say we improve the ensemble distribution function by enlarging the ensemble and then recalculate the effective property mean value and variance. What is the relation of these revised values to those determined with the more approximate ensemble distribution function?

Both the variances of the effective property values and the error owing to input uncertainty would be useful for defining bounds for the range of responses that could occur. Are the desired bounds related to those obtained by variational methods from an energy principle? In this regard it seems pertinent that the principle of complementary energy and the principle of minimum energy consider different BCs, but we are working with BCI values of effective properties. Also, the principles deal with energy definitions of effective properties, whereas the numerical analysis works with mechanically defined effective properties.

Beyond these questions of the applicability of variational bounds to effective property values determined by direct numerical simulation are other questions that bear on the variety of systems to which the variational methods can be applied. In particular is the applicability of variational bounding methods limited to perfectly bonded, linear materials lacking any discontinuities across internal interfaces? Do extremum principles exist to allow bounding methods to be applied to nonlinear or inelastic behavior? Do variational energy principles provide noncoincident bounds when the system size exceeds the RV? It seems likely that they do because system size does not enter into the bounding method, but why does system size not enter into variational bounding method?

5. Establishing Conditions for BC Independence

As stated at the end of Section 2, we presume that for a given composite, some aspect of the statistical description of the internal geometry of the material heterogeneities indicates or, perhaps, determines whether a volume yields BC-independent averages. Verifying that this is the case and identifying the particular feature of the statistical description of the geometry that is an indicator will require numerical experimentation with model composite systems. In this section we discuss items related to such an investigation. Particular investigations that will be pursued are outlined in Section 7.

The numerical investigation on model systems will evaluate the suitability of various statistical descriptions of the internal configuration of the material. For a simple material, like a uniform matrix with spherical inclusions, the pair distribution function, $g(\underline{r})$, or radial distribution function, $g(r)$, may be sufficient to statistically characterize the material. $g(r)dr$ is the probability of finding an inclusion whose center lies within an annulus, centered on a given inclusion, that has a thickness dr and radius of r. For slightly more complicated materials, like short fibers in a uniform matrix or micro-cracks in an otherwise homogeneous medium, the pair distribution function could be generalized to a vector function that additionally accounts for inclusion shape and orientation. Pyrz suggests using the second order intensity function, $K(r)$, because it is a theoretically well-understood mathematical quantity;[20] its radial derivative equals $2\pi r g(r)$. Pyrz demonstrated that $K(r)$ can discriminate between different patterns.

Statistical homogeneity (see Section 2) is likely to be a sufficient condition for a volume to have BC-independent average properties. However, it is an overly restrictive requirement for a given sized volume to be accepted as the RV size. Requiring statistical homogeneity for the RV needlessly dissolves the distinction between "statistically representative" and "statistically homogeneous." Instead of requiring that the RV be statistically homogeneous, it seems reasonable to expect that some aspect of the statistical description of the internal geometry should indicate the size for which averages become nearly BC independent. For example, it may happen that a distribution function chosen as the statistical description of a material changes qualitatively, but continuously, as the averaging volume is increased sufficiently to provide BCI. Such an occurrence could be likened to a phase transition, but one occurring in the distribution function rather than in the physical system. This opens the possibility of applying the analysis techniques used for phase transitions to try to identify, directly, the size of the RV. Such an analysis would be additionally attractive if the parameter that describes the transition in the statistical distribution (the order parameter) can be related to the physical system. As background for anticipated developments along these lines, the next section presents the physical interpretation of the correlation length and a brief overview of current understanding of continuous phase transitions.

6. Critical Phenomena and Correlation Length

The correlation length is a low-order measure of a statistical distribution that, owing to its physical significance, is a potentially useful quantity to use to express the criteria that the internal geometry of a heterogeneous material must meet for volume averages to be nearly BC independent. The RV and effective properties determined from it are only of interest when a low-order approximation for the response of a material is adequate. For this reason it is consistent to consider using a low-order approximation to the statistical distribution for the purpose of identifying the size of the RV. To illustrate the physical meaning of the correlation length, basic aspects of critical phenomena and the theory describing it are presented. This section also provides background for understanding several lines of inquiry current in the literature in which the behavior of disordered systems are represented by a variety of stochastic models. For this purpose brief descriptions of the scaling hypoth-

esis and percolation theory are presented along with a discussion of the (doubtful) relevance of critical phenomena to micromechanical modeling.

"Critical phenomena" is the modern term for the behavior previously referred to as "higher-order phase transitions" or "continuous phase transitions." "Phenomena," especially in the current discussion, should be understood in a very general sense, not limited to thermodynamic phase transformations. A system exhibiting a critical behavior undergoes a continuous, qualitative change in its character. Some attribute of the system changes continuously from zero, when the temperature, or some other control variable, is above a threshold value, to nonzero and growing for temperatures below the threshold. The quantity that changes continuously from zero is called the "order parameter."[21,22] While the order parameter is zero, the system is entirely in the high-temperature, or "disordered," phase. Nonzero values of the order parameter indicate that the system is a mixture of ordered and disordered phases. The magnitude of the order parameter provides a measure of the amount of the ordered phase present in the system. (The nature of order parameters is such that the new phase that is present only when an order parameter is nonzero has lower symmetry or can be construed to be the more ordered phase.)

Continuous thermodynamic phase transitions are the prototypical critical phenomena, with two cases being particularly accessible examples:[21] (i) liquid-vapor transition in a gas at its critical point (CP), which is the point in the pressure-temperature plane where the first-order liquid-gas phase boundary ends and (ii) ferromagnetic-paramagnetic transition in a ferrous metal at the Curie temperature. The first distinguishing feature of these transitions is that specific volume or magnetization, which are first derivatives of the Gibbs free energy (with respect to pressure or magnetic field, respectively), are continuous across the respective transitions. The order parameters of these two transformations are the difference in density between the vapor and liquid and the net magnetization. Careful observation demonstrates that the derivatives of the order parameter with respect to the intensive field variables (which are second derivatives of the Gibbs energy) are not just discontinuous but also have singularities at the critical point.

The concept of correlation length is motivated by the physical significance of these singularities. The pressure derivative of the density is the compressibility and the magnetic field derivative of the magnetization is the magnetic susceptibility. Near the Curie temperature the latter becomes very large and large magnetization fluctuations result from very small magnetic field fluctuations. Near the liquid-vapor CP, the compressibility becomes very large, and large density fluctuations result from very small pressure fluctuations. Experimental confirmation of the large density fluctuations comes from observations that the usually transparent fluid becomes milky white and opaque very near to the CP. This "critical opalescence," as it is called, results from density fluctuations acquiring sufficient magnitudes at wavelengths comparable to visible light that they strongly scatter light. In general, the derivative of an extensive thermodynamic variable with respect to the related intensive variable (a "thermodynamic force") is referred to as a "response function" or "susceptibility."[9,22] Elastic constants are a notable example of such quantities. These susceptibilities are the quantities that become large near a CP and singular at it. Consideration of thermodynamic fluctuations in a system at thermodynamic equilibrium leads to the conclusion that the susceptibilities govern the spatial extent of fluctuations of the extensive

variables.[21,22] In particular in the thermodynamic limit, the static (zero frequency) susceptibility is completely determined by the long wavelength, equilibrium fluctuations.[9] Hence divergence of a susceptibility indicates that the extent of the corresponding fluctuations becomes as large as the system itself. The existence of long wavelength fluctuations means that the response of the system is correlated over macroscopic distances. There is long-ranged order. The order parameter value at one location becomes related to its value at a distant location. Indeed, all intensive quantities acquire long-range correlations near a CP, as explained next.

Thus critical phenomena depend on fluctuations in an essential way. Far from a CP, fluctuations are of microscopic extent, comparable to the interparticle separation; but as the CP is approached, they occur on an increasingly broad spectrum of wavelengths. At the CP fluctuations at all wavelengths - *i.e.,* all length scales - contribute to the values of the thermodynamic variables.[9,23,24] Mathematically fluctuations are described by the correlation function. The long wavelength fluctuations correspond to the mean of the correlation function.[9] The correlation length is a characteristic length associated with the correlation function. As the susceptibilities grow near the CP, the correlation function and its correlation length also diverge. This correspondence indicates that the correlation length, ξ, provides a measure of the spatial range of order in the system. In particular the correlation length marks the *crossover* between critical and ordinary behaviors:[24] When a system is treated on a length scale, L, - *i.e.,* averaged over a region of size L^d, where d is the system dimensionality - for which $L \ll \xi$, it exhibits power law critical behavior, just as at the CP. In contrast on length scales with $L \gg \xi$, correlations decay exponentially fast and the behavior is ordinary.

The correlation between two (or more) events is the difference between the joint probability of the pair of events, P_2, and the product of the random probability of each event alone, P_1.[23] A useful example is the pair of events in a system of particles that there is a particle located at point A, and another particle located at point B such that the distance vector between these two positions is $\vec{r}$. The correlation, as a function of $\vec{r}$, is given by

$$C(\vec{r}) = \frac{1}{2}P_2(\vec{r}) - P_1(\vec{r_A})P_1(\vec{r_B}) . \qquad \textbf{(12)}$$

To make this relation statistically meaningful, it needs to be averaged over an ensemble of similar systems. The ensemble average is typically equated with a time average or a space average over a single macroscopic sample by invoking ergodicity.

To extend this example, consider a simple lattice model of a binary alloy that entails only very short-ranged interactions. Investigations of such models show that the state of occupation of a substituted lattice is characterized by a wide-spread order even though the interactions only have a short range.[23] For understanding how such long-ranged effects arise

it is useful to study the correlation function, $C(\vec{r})$. Theories of critical phenomena predict a general form for the radial dependence of the correlation function given by[23]

$$C(r) \sim r^{-n} \exp(-r/\xi) . \quad \textbf{(13)}$$

Based on this behavior, the correlation length is defined as[25]

$$\xi^{-1} = \lim_{r \to \infty} \frac{-1}{C} \frac{\partial C}{\partial r} . \quad \textbf{(14)}$$

Hence ξ has the physical significance of being a spatial range of correlations within the system.[23]An alternate scalar measure of correlations that gives the range of order is

$$\Gamma^2 = \int r^2 C(|r|) d^3 r / \int C(|r|) d^3 r . \quad \textbf{(15)}$$

At the CP the correlation length, ξ, and range of order, Γ, both become infinite.

6.1 Universality and the Scaling Hypothesis

That fluctuations at all length scales contribute to the values of the thermodynamic variables at the CP means that there is a loss of length scale for the system. The discrete lattice spacing becomes irrelevant, and there is no fluctuation wavelength with any special significance. Another manifestation of the loss of scale is that based on measurement and analysis of model systems, the (singular part of the) thermodynamic functions are described by power laws in the distance to the CP.[9,21] Power laws are scale invariant, meaning that the form of the function is unchanged by multiplying its argument by a scale factor. Consequently, the function is self-similar at all scales. It has no characteristic length associated with it.[26] Basic thermodynamic and statistical mechanical arguments show that the exponents of these power laws, the "critical exponents," satisfy a set of inequality relations, independent of the details of any particular system.[21] This is consistent with the idea that critical phenomena arise from long-ranged correlations, for which case it is reasonable to expect that at least some details of the interactions within the system are irrelevant to the behavior near the CP.[23] Accepting this assumption, systems that share the same relevant aspects would be expected to have comparable singular behavior at the CP. These expectations have lead to the universality hypothesis, which asserts that all critical behaviors may be classified according to the dimensionality of the system, the symmetry group of the order parameter, and possibly a few other general criteria.[23,25] For this reason it is of considerable interest to determine critical exponents of the different universality classes. Knowing these, the dimensionality of a system, and the symmetry of its order parameter so that its universality class can be identified, the qualitative behavior of the system near a CP is known, if the universality hypothesis is correct.

The value of being able to describe thermodynamic functions near the CP and the inference that a loss of length scale is intrinsic to critical behavior motivated the scaling hy-

pothesis.[9,21,23-25] At the CP the correlation length, ξ, which marks the crossover between critical and ordinary behaviors, diverges, making all finite lengths that are representative of aspects of the system much smaller than ξ. For this reason, at the CP it should not matter what length scale is used in investigating the system; the system looks similar at all length scales from the lattice spacing up to the system size. Based on these observations, Wilson advanced the scaling hypothesis: As a CP is approached, thermodynamic functions change their scale but not their functional form. This is described mathematically by expressing the thermodynamic free energy as a homogeneous function (a generalized power law).[9,21] Scaling theory, which is the result of taking any free energy to be a homogeneous function in the neighborhood of a CP, leads to a consistent description of the critical behavior. This result shows the scaling hypothesis to be a unifying principle for critical phenomena. Beyond this scaling theory produces *equality* relations among the critical exponents and constrains the form of the equation of state. Most notably scaling theory predicts that only two critical exponents are independent. The predicted scaling behavior of thermodynamic functions near a CP has been partially verified experimentally in addition to analytic and numerical verification for model systems. Scaling theory does not predict the values of the critical exponents, however. For determining these, a set of scale transformation techniques, collectively known as renormalizaton group methods, have been developed.[25]

6.2 Percolation

Percolation is a purely mathematical procedure that exhibits a kind of critical behavior that is not a thermodynamic phase transition nor even a physical phenomenon.[24,27] It has been used to good advantage as a technical tool to model a broad range of physical transition phenomena.[27] It was originally introduced as a model of fluid flow in a disordered medium and recently has been investigated extensively as a model for fracture in disordered materials.[28,29] The latter is particularly interesting in the context of the present inquiry, as indicated in the introduction. We discuss percolation here as background for the anticipated future investigation of microstructurally based fracture criteria. In addition, percolation provides further illustration of the physical meaning of the correlation length. That percolation theory provides a richer understanding of correlation length is an example of why it has attracted much attention. Generally speaking, the critical behavior exhibited by percolation models is highly accessible to investigation. Combined with the universality hypothesis, this makes percolation a useful model of critical phenomena in physical systems. Investigation of a percolation system having the same order parameter dimensionality and the same spatial dimensionality as a physical system of interest can provide an accurate description of the near-CP response of the physical system if the universality hypothesis is correct. Furthermore, percolation can provide insight into localized-to-extended state transitions when applied as a qualitative model of the physical phenomenon.[27]

In lattice percolation one considers randomly filling (marking) sites or bonds of a regular lattice with some fixed probability, p.[24,27] The *statistical* question of interest is, 'for what value of the filling probability, *on average*, does a continuous path of marked sites first develop through an arbitrarily large lattice?' This critical fraction of filled sites is the

"percolation threshold," p_c ,and it is a critical point.[24] The most noteworthy feature of the percolation problem is that it is a simple model that exhibits critical phenomena. In this case the "phase transition" is geometric, going from the absence of any continuous paths spanning the entire system to the existence of such a path, which is called the "infinite cluster." The infinite cluster covers an increasing portion of the lattice as the probability of occupancy of the sites of the lattice is increased. The order parameter is the fraction of the system occupied by the infinite cluster. Because lattice percolation and its generalization to continuous space, continuum percolation, can be conveniently investigated numerically, they are useful for modeling. Furthermore, the details of systems that exhibit percolation are quite varied. Only the simplest site percolation systems have been alluded to here. The flexibility in defining percolation systems is an additional reason for their growing use for modeling physical systems. Bond percolation is another class of percolation problems that exhibit distinct differences from site percolation.

It should be emphasized that percolation is a stochastic model. This is typical of critical phenomena, which are governed by system fluctuations and, consequently, dependent on the statistical distribution of variable values, not just on their mean values.[27] The procedure for investigating percolation numerically is to randomly fill a collection of identical finite-sized lattices using a given filling probability. This process generates a large number of individual cases or "realizations" of the same overall state - namely, a fixed mean value of the fraction of filled sites. This collection of many realizations is treated as an ensemble of configurations, and conclusions concerning the behavior of a system that exhibits percolation are statistical statements about the behavior of an ensemble of realizations of the system subject to prescribed values of its independent controlling variables. This is the reason for emphasizing "on average" in describing the percolation problem. For systems whose analytic description is sufficiently simple, the alternative to working with an ensemble of systems is to explicitly determine the behavior of one realization of the system in the thermodynamic, infinite size limit.

The range of order, Eq. (15), is used in percolation theory as an alternate definition for the correlation length.[24,25] In the context of percolation, the correlation function is the probability that a site a distance, r , from an occupied site is also occupied and is in the same cluster. (A cluster is the group of all the sites connected together by a continuous path of neighboring occupied sites.) The square of the range of order is then

$$\Gamma^2 = \frac{\sum' r^2 C(r)}{\sum' C(r)}, \qquad \textbf{(16)}$$

where the summations run over all lattice sites. Γ^2 determined in this way is the average squared distance between two cluster sites in the lattice. Γ is consistent with that for thermodynamic phase transitions, Eq. (6), in as much as the critical exponent for it satisfies the expected relations with the other exponents.[25] Γ is also a measure of the largest hole in a cluster and the radius of those clusters that make the main contribution to the mean cluster size.[24]

The physical significance of the correlation length and scaling properties of percolating systems were further illustrated by a numerical study of site percolation on a two-dimensional square lattice.[30] Kapitulnik *et al.* demonstrated that near the percolation threshold, the infinite cluster is homogeneous on length scales much larger than ξ and ramified with holes on all scales smaller than ξ. Thus ξ is a measure of the size of heterogeneities in the lattice.

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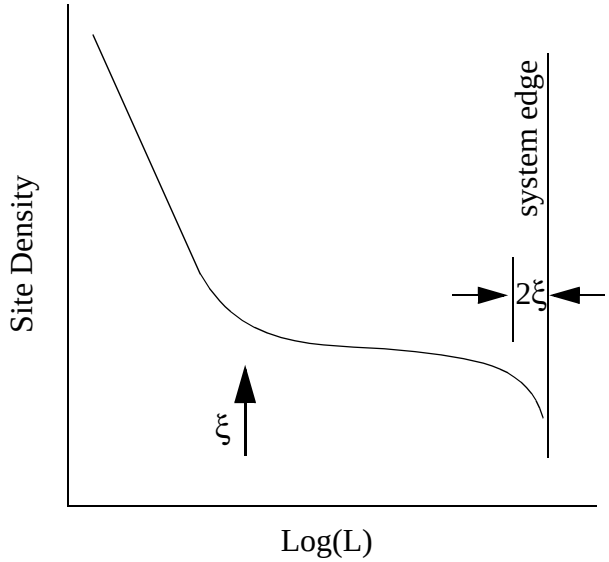

Figure 1. Schematic semilog plot of site density versus linear dimension of averaging window for site percolation on a square lattice (after Kapitulnik *et al.*[30]).

Figure 1 is a schematic of their results. For averaging areas L^2 within the system with $L < \xi$, the density of occupied sites in the cluster varies as a power of L. This is indicated by the linear part of the curve at small L. For averaging areas with $L > \xi$, the cluster density is roughly constant and the system appears to be homogeneous. This corresponds to the shallow-sloping midportion of the curve. Their study also demonstrated a further effect of correlations. As the averaging area approached the size of the simulated system, the calculated cluster density again varied with the size of the averaging area, as indicated by the steepening of the curve at large L. This result is an example of calculated values of system properties being sensitive to the boundary conditions. Appropriately the range of influence of these boundary effects is given by the correlation length, ξ. This is consistent with the notion that the correlation length characterizes the range of influence between separate regions of the system.

By repeating the numerical exercise for a different value of p, the probability of site occupancy, Kapitulnik *et al.* also found that the percolation system exhibited scaling behavior.[30] They showed that ξ increased as p neared p_c, and the average site density on the homogeneous scale ($L > \xi$) decreased, both in accordance with the expected power law behavior. Far below the percolation threshold, only small clusters occur and the correlation

length is on the order of the lattice spacing, a . In this regime the lattice spacing provides a relevant length scale. Near the percolation threshold the lattice spacing becomes unimportant because the range of cluster sizes increases substantially, and the clusters contain holes of all sizes smaller than themselves. The resulting ramification of the clusters corresponds, for critical phase transitions, to the thermodynamic functions being determined by fluctuations with an increasingly wide range of wavelengths. In that same sense there is a loss of length scale. The correlation length, which can be interpreted as the radius of those clusters that make the dominant contribution to the mean cluster size,[24] is the only available length scale. Both it and the size of the infinite cluster, which is the order parameter, diverge as power laws near the percolation threshold, p_c , as is expected from the scaling hypothesis.

Generally speaking, ξ can be a measure of heterogeneity. In percolation it gives the linear extent of geometric heterogeneities. In physical systems it characterizes the linear extent of heterogeneities in field variable values. Because these are the attributes that we desire for an RV, the correlation length is an appealing candidate for defining RV size. Several questions need to be resolved to establish that a correlation length is suitable for the RV size. When applied to a mechanical system, does the correlation length retain all of the attributes that it has for critical phenomena? What is the analogy to critical phenomena? What correlations are of interest? How should an RV defined in terms of a correlation length be used? Does scaling behavior occur? Under what conditions? The discussion in the next subsection bears on these questions. Answers to these questions will be pursued through the numerical experiments on model heterogeneous systems that are described in Section 7.

6.3 Discussion

A key question to resolve in determining how to use the numerical simulations of microstructural processes to develop constitutive models is whether real microstructural mechanics processes exhibit critical phenomena. A micromechanical system that does have critical behavior is highly amenable to investigation with a percolation model. In addition, we can immediately infer several qualitative aspects of the behavior of a system in which critical microstructural processes occur: The “interesting” macroscopic behavior of the system is governed by the distribution of values of the microstructural quantities, not just their mean values.[23,27] A correlation length, ξ, can be defined that characterizes the statistical distribution, which evolves during deformation. The correlation length could be taken to define the linear dimension of an RV because it marks the crossover between small length scales, on which different regions of the system behave differently, and large length scales, on which all regions of the system are comparable. Presumably the requirements of EMT are satisfied for RVs of this size subjected to macroscopically uniform BCs. Provided the mesh elements used in a FEM simulation are larger than ξ, the material is macroscopically homogeneous.

Near the CP the correlation length is finite but becomes macroscopically large. Thus if a micromechanical system exhibits critical behavior, the RV has a variable size, and near the CP the RV can exceed the size of mesh elements. When this occurs, the material within one mesh element may not be equivalent to the material within another element. A consti-

tutive description need not be abandoned, but a different constitutive model has to now be used to the extent that well-defined constitutive variables cannot be determined by simple volume averaging over an RV. The material response may become nonlocal. Where RV averaging is inadequate, an alternative approach to defining relevant constitutive quantities may present itself. Because the system is near a CP, applying the scaling hypothesis may provide a link between microstructural and macroscopic response and may be a basis for developing an appropriately altered constitutive model. This last possibility needs to be investigated more fully.

A related question is whether a numerical model chosen to represent regular (noncritical) micromechanical behavior can, itself, exhibit critical behavior. When this is a possibility, the region of parameter space that produces critical behavior in the numerical model then needs to be identified and avoided when the model is applied.

Preliminary to determining whether a given micromechanical system or numerical model exhibits critical behavior, it will be necessary to develop a statistical description of a material's heterogeneity. A useful statistical description will represent the distribution of the variables whose correlations control the material response. Such a statistical description is desirable, regardless of whether there is critical behavior, as discussed in Section 5.

In order to apply a typical percolation model to represent microcracking behavior, several questions should be considered. Is the ultimate state of pervasive cracking that produces a system-spanning fracture critical behavior, as is implicit in the percolation modeling approach? The first step required to answer this question is to identify the microstructural quantity whose fluctuations exert a significant influence on the macroscopic behavior. Does this quantity qualify as an order parameter? Large values should correspond to being far from the CP, and it should be discontinuous across a first-order transition with the discontinuity vanishing as the CP is approached.[23] Second, do these fluctuations become critical - that is, does their correlation length diverge and are there fluctuations at all wavelengths when it does? (Together, these indicate a loss of length scale.) Answering these questions not only identifies whether we are dealing with critical phenomena but also establishes the analogy to a model critical system. This is important for interpreting the results when, *e.g.*, a percolation model is used to represent microcracking resulting in fracture.

Disorder is cited as a defining feature of the systems in which microcracking has been modeled with percolation.[28] The disorder can be of any kind, from extensive material heterogeneity, or just local strength variability within a single crystal. Some aspects of the response of disordered systems can be modeled as critical phenomena when the statistical distribution of the disorder, not simply the average amount of disorder, governs the system behavior. Recall that the spread of the probability distribution of the disorder can be characterized by a correlation function. Hence the correlation length can provide a measure of the fluctuations of the spatial disorder within an ensemble of systems. When the response depends on only the average amount of the disorder, its analysis does not require ensemble averaging or, equivalently, taking thermodynamic limits.

The elastic network is a version of a bond percolation model that has been investigated in an attempt to obtain insight into cracking in disordered brittle materials. The situation

that is modeled by an elastic network is microcrack nucleation at randomly located flaws or weaknesses and coalescence to form a macroscopic fracture in an initially uniform body subjected to far-field tension.[28] It has also been investigated with some experimental percolation models.[31,32] In contrast to a typical percolation model, evolution of the elastic network is more directly governed by physics. For each increment in strain, the stresses in the network of elastic bonds are explicitly calculated. The bond that reaches a failure condition first is removed and the stresses are recalculated. This is repeated for a given strain increment until equilibrated stresses are obtained for which no more bonds reach failure. In further contrast with typical percolation models, the resulting ensemble behavior of the elastic network systems has been interpreted as exhibiting a scaling behavior for typical situations and not the scaling that develops near a CP.[33] The noncritical scaling is likened to the way the scaling behavior of turbulence is described.[33] The concept of noncritical scaling and the role that a correlation length may play is not yet clear and needs to be studied further.

7. Initial Study

To begin to investigate the issues that have been identified, in particular criteria for defining RV size, we will pursue the following investigations.

To illustrate the concepts of macroscopically equivalent systems and bulk property determination in statistical physics, we will investigate the calculation of stress in a discrete system using molecular dynamics simulations. The two parts of Eq. (A4), presented in the Appendix, will be evaluated for model systems to verify the points stated by Balescu. The model systems will be an FCC crystal of atoms interacting according to a Lennard-Jones potential and a similar FCC crystal having randomly positioned point defects. The perfect crystal will be treated at finite temperature, where thermal motion will introduce irregularity. The defective crystal will be analyzed at zero temperature after the system is allowed to relax to an equilibrium configuration. For both types of systems, the simulations will proceed by deforming a large system to induce a state of stress. This will be followed by calculation of the bulk and surface contributions to macroscopic stress from the two parts of Eq. (A4) for a variety of subvolumes.

The results of these calculations will be evaluated for the expected trends given by Eqs. (1) and (2) and to distinguish system surface effects from averaging region size effects. The latter evaluations will entail comparison of distances within the system with a relevant correlation length. For the crystal with point defects, this should be related to the radial distribution function of the point defects. For the perfect crystal the minimum-sized bulk system is the unit cell at zero temperature. For a warm crystal it is larger with the relevant correlation length expected to be related to the wavelength of some characteristic lattice vibration.

Another investigation will assess the criteria for defining the RV discussed in the present report. In the first part of this study, we will seek to confirm that equilibrium effective properties that are independent of the choice of BCs can be obtained by increasing the system volume and/or the averaging volume for a heterogeneous continuum. The averaging volume need not be as large as the system volume. As discussed above, in the case of site percolation the averaging volume needed to be smaller than the system volume to avoid

boundary effects.[30] Two heterogeneous continuum models are needed to perform this exercise. The first should maintain continuity throughout the deformation so that the fundamental results of effective medium theory can be confirmed for numerical application. The second model should include some form of cracking to test our ability to properly treat discontinuities in otherwise continuous systems. Suitable microstructural test models will be sought from those that are being developed and applied in concurrent projects at SNL, both for expediency and because it is anticipated that these two projects will be able to use the results of the present project. Candidate model systems to use in these investigations include: glass microballoon-filled epoxy, an elastically and plastically deformable polycrystal aggregate, and a randomly microcracked elastic body.

The second part of this study is to address how to statistically characterize the heterogeneity and identify aspects of the statistical characterization that indicates RVE size. This will be pursued by testing different means of computing and then interpreting correlation lengths for all of the simulations performed in the first part of this study. Developing algorithms for computing the correlation function and length in the types of numerical models of interest is a necessary first step. Following this will be the more interesting step of interpreting the resulting correlation information. This will be aimed at relating the correlation length of the most relevant variables to criteria for BCI of equilibrium effective properties.

It will be of interest to follow up this assessment of criteria for identifying the RV by using the results of the above investigation to determine the extent to which fracturing can be regarded as a critical phenomenon. It would be appropriate for this purpose to use an elastic network model in which fracture has been treated by others as being a critical phenomenon.[28] The computer program GLAD, being used in two projects at SNL is such a model. This makes a close comparison with previous investigations more manageable. Given our ambition of developing criteria relating microcracking to macroscopic cracking, it will be useful to clearly identify which aspects of microscopic cracking can be associated with critical behavior. Under conditions that elicit the supposed critical behavior of microcracking, the relation to macrocracking must change. Volume averaging should become untenable owing to correlation lengths enlarging, but at the same time, scaling relations should become applicable. Thus this part of the study, seeking to identify critical behavior in a model system with cracking, is the first step in properly treating the macroscopic consequences of possible critical behavior for microcracks.

Beyond these particular investigations, attention needs to be given to how to develop effective property treatments of inelastic (nonlinear) response and to identifying how to simplify the constitutive description in a simulation on a length scale smaller than the RVE.

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APPENDIX

Example Bulk Property Calculation: Stress in a Discrete System

The configurational stress in a system of particles whose interaction is described by the crystal potential, Φ, is given by[a1]

$$\boldsymbol{\sigma}_{ij} = \frac{1}{V}\sum_{\alpha=1}^{N} R_i^{\alpha}\nabla_j^{\alpha}\Phi \ , \qquad \textbf{(A1)}$$

where R_i^{α} is the i$^{\text{th}}$ component of the position vector to the α^{th} particle. Greek superscripts label particles and latin indices label Cartesian components. Consider the case where the crystal potential is the sum of pairwise interactions between the particles. We have

$$\Phi = \sum_{\alpha=1}^{N}\sum_{\beta=\alpha+1}^{N}\phi^{\alpha\beta} = \frac{1}{2}\sum_{\alpha,\beta=1}^{N}\phi^{\alpha\beta}, \qquad \textbf{(A2)}$$

and taking $R_i^{\alpha\beta} \equiv R_i^{\alpha} - R_i^{\beta}$, the configurational stress becomes

$$\sigma_{ij} = \frac{1}{2V}\sum_{\alpha,\beta=1}^{N} R_i^{\alpha\beta}\nabla_j^{\alpha\beta}\phi^{\alpha\beta} = \frac{1}{2V}\sum_{\alpha,\beta=1}^{N}\frac{R_i^{\alpha\beta}R_j^{\alpha\beta}}{\left|\vec{R}^{\alpha\beta}\right|}\frac{\partial\phi^{\alpha\beta}}{\partial\left|\vec{R}^{\alpha\beta}\right|} \ . \qquad \textbf{(A3)}$$

Assuming that the particle interactions have a finite range, this expression can be separated into sums over M interior particles and $N-M$ near-surface particles results in a form similar to that of Eq. (1):

$$\sigma_{ij} = \frac{1}{2V}\sum_{\alpha=1}^{M}\sum_{\beta=1}^{N} R_i^{\alpha\beta}\nabla_j^{\alpha\beta}\phi^{\alpha\beta} + \frac{1}{2V}\sum_{\alpha=M+1}^{N}\sum_{\beta=1}^{N} R_i^{\alpha\beta}\nabla_j^{\alpha\beta}\phi^{\alpha\beta} \ . \qquad \textbf{(A4)}$$

The first double sum includes all of the interactions of the M particles that are farther from the surface than the particle interaction range. It is expected to give the bulk value of stress in the system, analogous to Λ in Eq. (1). Based on Balescu's characterization presented in Section 3, it is expected that this interior bulk contribution will be constant for sufficiently large systems. The second double sum includes all of the particles that are close enough to the surface to interact with particles (in the case of applied mechanical contact forces) that are outside of the system. Note that the interactions with the external particles are not included in the second double sum since Φ is the crystal potential not the total potential.[a1] The surface contribution to σ_{ij} expressed by the second double sum obeys Eq. (2) since, with increasing system size, V increases faster than the number of contribution to this double sum.

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